

Rejection Under 35 U.S.C. 112, second paragraph

The Examiner has rejected claims 1, 2, and 16 under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

A. The Examiner asserted that claim 16 was indefinite for "merely reciting a use without any active, positive steps delimiting how [the] use is actually practiced. Applicants submit that because the claim has been cancelled, the rejection is moot and should be withdrawn.

B. The Examiner asserted that in claim 1 the expression, "numeric mean dimension", was not clear. To more particularly point out applicants invention, the expression "numeric mean dimension" has been changed to "average numeral dimension", which is the correct expression to indicate to one of skill in the art that the average size of at least 80% of the total number of particles will be less than 1 micron. Applicants submit that rejection should be withdrawn.

C. The Examiner asserted that claim 1 was vague because it failed to specify what "values" were envisioned. Applicants have amended the claim to more particularly point out that the values referred to are pH values. Applicants submit that rejection should be withdrawn.

D. The Examiner questioned whether claim 2 referred to "only 'cellulose' or a 'cellulose ester'. The claim has been amended to particularly point out a cellulose ester. Applicants submit that rejection should be withdrawn.

Rejection of the Claims Under 35 U.S.C. 103 (a)

The Examiner has rejected claims 1-17 under 35 U.S.C. 103(a) as being unpatentable over European Patent Application No. EP 0722980 to Bastioli et al. (hereinafter "Bastioli '980") in view of the journal article "Biodegradable blends of cellulose acetate and starch: ...", by Mayer et al. (hereinafter "Mayer"), and Japanese Unexamined Patent Laid Open Application No. JP 07102114 (hereinafter "JP '114"). Applicants respectfully traverse the rejection and submit that a proper *prima facie* case of obviousness has not been made.

The criteria and Examiner's burden for making a *prima facie* case of obviousness in accordance with MPEP Section 706.02(j) are as follows (emphasis and numbers added):

To establish a *prima facie* case of obviousness, three basic criteria must be met. (1) First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. (2) Second, there must be a reasonable expectation of success. (3) Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations. The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art and not based on applicant's disclosure. . . .

The initial burden is on the examiner to provide some suggestion of the desirability of doing what the inventor has done. "To support the conclusion that the claimed invention is directed to obvious subject matter, either the references must expressly or impliedly suggest the claimed invention or the examiner must present a convincing line of reasoning as to why the artisan would have found the claimed invention to have been obvious in light of the teachings of the references." . . .

Further, case law makes clear that the best defense against the subtle but powerful attraction of a hindsight-based obviousness analysis is rigorous application of the requirement for a showing of the teaching or motivation to combine prior art references, and that that such

references can be combined only if there is some suggestion or incentive to do so. In re Sang-Su Lee, 61 U.S.P.Q.2d 1430 (CAFC 2002).

Bastoli '980 In View Of Mayer And JP '114

With regard to Bastoli '980, the Examiner asserts that:

Bastoli [sic] discloses biodegradable compositions comprising (a) starch, (b) a cellulose ester or ether, (c) a plasticizer for starchy phase, cellulose and derivatives and (d) and compatibilizer agent (abstract). Starch and cellulose are present in a range from 1:90 to 90:1 by wt (p. 10, lines 31-33). Plasticizer is present from 5-40% by wt (p. 10, line 55 to p. 11, line 1). Compatibilizing agents described on page 2, lines 31-50 read on those encompassed by instant claims 9-11. Various articles can be moulded. (p.2, lines 20-22).

Bastoli does not mention an additive (of instant claim 1). (Paper 7, p.4).

Next, with regard to Mayer, the Examiner asserts that:

Mayer discloses biodegradable blends of [cellulose] acetate and starch. Degree of substitution of cellose acetate should be 2.5 or less (p.776, 3rd paragraph). In expensive [sic] are added to the blends. (Paper 7, p.4).

With regard to JP '114, the Examiner asserts that:

[JP '114] also discloses biodegradable composition containing cellulose ester, starch and plasticizer together with an additive like talc, calcium carbonate, magnesium carbonate etc. (Paper 7, p.4).

Finally, the Examiner concludes by asserting that:

Therefore it would have been obvious to add to the composition of Bastoli [sic], carbonates and hydroxides of calcium and magnesium in order to reduce the cost of production, without affecting quality and performance. It would also have been obvious to use cellulose ester i.e. polysaccharide of degree of substitution of not more than 2.5 in order to maintain good biodegradability of the composition. (Paper 7, p.4).

The Examiner appears to be reasoning that it would have been obvious to add calcium carbonate or magnesium carbonate to the compositions of Bastoli '980 to reduce the cost of production of those compositions without affecting "quality" and "performance" because calcium carbonate and magnesium carbonate are mentioned as possible additives (together with

numerous other additives that are totally different from each other and from calcium and magnesium carbonates) in JP '114 for improving "[v]arious aspects of performance, starting with mechanical properties such as strength..." of the particular compositions disclosed in JP '114.

Applicants respectfully submit that the Examiner has not made a proper prima facie case of obviousness because one of skill in the art would have had no motivation to combine the disclosures in the manner the Examiner proposes.

The Examiner fails to appreciate that: 1) the function of the additive in the present invention and in JP '114 is completely different; 2) that the problem of increasing biodegradability of the claimed compositions, the compositions of Bastioli '980, and the compositions of JP '114 is achieved by different methods; and 3) that the amounts and ratios of cellulose ester, starch, and plasticizer for the compositions of the present invention compared to those of JP '114 differ so greatly that one of skill in the art would not be motivated to modify the present invention with the additives of JP '114. A complete English language translation of JP '114 is provided as Exhibit 2.

First, Applicants point out that there is no teaching or suggestion anywhere in JP '114 of the possible role of the additives regulating pH to improve the biodegradability of the compositions. Instead, the disclosure of JP '114 focuses on the addition of additives for improving strength. Of all the additives JP '114 mentions, "talc, calcium carbonate, magnesium carbonate, clay, silica, alumina, glass powder, mica, wood, cellulose, polyvinyl alcohol, chitin, chitosan, collagen, fibroin, and keratin", only calcium and magnesium carbonate and clay, are effective at increasing the biodegradability of the compositions of the present invention (i.e., starch dispersed in a matrix of cellulose acetate as claimed). Because JP '114 teaches a different use, and because the vast majority of the additives listed in JP '114 could not meet the

requirements of the claimed compositions, it would not have been obvious for one of skill in the art to choose calcium and magnesium carbonate from the large list of additives in JP' 114 to combine with the claimed compositions. Moreover, even if there was such motivation, nothing in either Bastioli '980 or JP' 114 would provide a person of ordinary skill in the art with an expectation that adding a "pH-raiser" as an additive would increase the biodegradability of the composition. For these reasons, applicants respectfully submit that the rejection should be withdrawn.

Next, applicants point out that the compositions described in Bastioli '980 are endowed with an improved biodegradability thanks to the high degree of dispersion of starch in the cellulose acetate matrix as conferred by the specific phase agents therein used. However, despite the high dispersion of starch, the decomposition rate during composting of the articles produced from those compositions is still slow compared to the present invention (specification, p. 1, last two paragraphs; and comparison examples 1-4).

JP '114 addresses the problem of increasing the biodegradability of the starch-cellulose acetate compositions by increasing the content of plasticizer of the compositions, "the amount of plasticizer used must be from 30% to 70% wt with respect to the total weight of the composition which includes the cellulose ester, starch, and plasticizer" (Translation of JP '114, p. 4, final paragraph).

Examination of JP '114, Example 1, experiments 2 and 3, see Table 1, shows that only when plasticizer is used in a proportion of 55% and 63% to the weight of starch, cellulose diacetate, and plasticizer, are the articles produced almost completely degraded after 3 months of being embedded in adequately humid soil (Translation of JP '114, p. 6, para. 0031). When the plasticizer is used in a proportion of 47% and 33%, Table 1, experiments 1 and 6, the

biodegradability is reduced. Similarly, when the plasticizer is used in a proportion of 33% and 41 %, Table 2, experiments 7-13, the biodegradability is even worse (part of the filament was undamaged).

In the claimed compositions, the quantity of plasticizer is from 10 to 40 wt% of the plasticized cellulose ester (i.e. the sum of the ester and the plasticizer). Taking into consideration, that the amount of plasticizer used in the compositions of Bastioli '980 is only from 5 to 40 wt% of the total composition, one of skill in the art would see that this is below the 55% proportion of plasticizer shown to be necessary in the compositions of JP '114 for achieving near full biodegradability. Therefore, one of ordinary skill in the art would have not considered the compositions of Bastioli '980 as being suitable candidates for addition of the additives of JP '114, let alone calcium and/or magnesium carbonate specifically, because there is no evidence that a biodegradable composition as claimed would have been produced. For this additional reason, the rejection should be withdrawn.

Finally, applicants point out that there are critical differences in the amounts and ratios of starch, cellulose ester, and plasticizer of the claimed compositions and those of JP '114 that would dissuade one of skill in the art from believing that Bastioli '980 could and should be modified with the additives of JP '114. As pointed out above, JP '114 describes biodegradable compositions containing a cellulose ester, starch, and plasticizer where an essential requirement of the compositions is that "the amount of plasticizer used must be from 30% to 70% wt with respect to the total weight of the composition which includes the cellulose ester, starch, and plasticizer" (Translation of JP '114, p. 4, final paragraph). In other words, the compositions of Kokai '114 must comply with the following relationship:

- (1) $P/(E+A+P) = 0.3$ to 0.7 , where hereinafter

P = plasticizer

A = starch

E = cellulose ester

Also, to avoid a "biodegradation rate that is too slow and the material is impractical" as well as a where the "moulding [sic] properties [are] adversely affected, [and] the biodegradation rate is too high and the composition is unstable" (Translation of JP '114, p. 4, first and second full paragraph), the ratio by weight of cellulose ester and starch must be from 99:1 to 20:80, which may be expressed as:

- (2) $99:1 > E:A > 20:80$.

On the other hand, for the claimed compositions (see claim 1), the ratio by weight between starch and plasticized polysaccharide ester is from 1:6 to 1:18, that is:

- (3) $1:0.6 < A:(E+P) < 1:18$;

Also, the polysaccharide ester is plasticized with a plasticizer in a quantity of from 10 to 40% by wt. referred to the polysaccharide ester (see claim 1), that is:

- (4) $0.1 < P/E < 0.4$;

Starting from equations (3) and (4) above, one may calculate the values of $P/(E+A+P)$ for the claimed compositions, so that they may be compared to those of JP '114 on a common basis. Please see attached calculation sheets (Exhibit 3) for full solutions. The following values are obtained:

$P/(E+A+P) = 3.4\%$ to 27% for the claimed compositions, which is below the values of 30 to 70% required by JP '114.

In the same manner, one may start from equations (1) and (2) for JP '114, and calculate the ratio P/E for a comparison to the claimed compositions. Again, please see attached calculation sheets for a full solution. The following results are obtained:

$P/E = 43$ to 1165% for the compositions of JP '114, which is much higher than the values of 10 to 40% of the claimed compositions.

A graphical comparison of the solutions for the claimed compositions and the compositions of JP '114 is also provided with the calculation sheets (See Exhibit 3).

These significant differences in the characterizing features of the claimed invention and JP '114 relating to the amount of the plasticizer and relative amounts of starch and cellulose ester demonstrate that the compositions are very different. Thus, one of skill in the art would not reasonably believe that the additives used in one set of compositions (JP '114) could successfully be used to modify another set of compositions (the presently claimed invention), especially where the principals for achieving biodegradability of the two compositions are completely different.

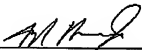
Because it has shown above that: 1) the function of the additive in the present invention and in JP '114 is completely different; 2) the problem of increasing biodegradability of the claimed compositions, the compositions of Bastioli '980, and the compositions of JP '114 is achieved by different methods; and 3) the amounts and ratios of cellulose ester, starch, and plasticizer for the compositions of the present invention compared to those of JP '114 differ so greatly that one of skill in the art would not be motivated to modify the present invention with

the additives of JP '114, applicants respectfully submit that a proper prima facie case of obviousness has not been made and the rejection must be withdrawn.

CONCLUSION

In view of the foregoing, favorable action on the merits, including withdrawal of the rejections, and allowance of all the claims, is respectfully requested. If the Examiner has any questions regarding this paper, please contact one of the undersigned attorneys.

I hereby certify that this correspondence is being deposited with the United States Postal Service as first class mail in an envelope addressed to the Commissioner for Patents, Washington, D.C. 20231, on September 20, 2002.



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EXHIBIT 1

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ABSTRACTBiodegradable compositions comprising starch and polysaccharide esters

C1
The biodegradable heterophase compositions comprise partially or completely destructured and/or complexed starch, a polysaccharide ester and a plasticizer for the polysaccharide ester. In these compositions, the polysaccharide ester constitutes the matrix and the starch the dispersed phase in the form of particles or domains of numeric mean dimension lower than 1 μm and preferably than 0.5 μm for at least 80% of the particles. The compositions also comprise an additive which can increase and maintain at values of 4 or more the pH of a solution obtained by placing the compositions in pellet form in contact with water at ambient temperature for 1 hour, with the use of a pellet:water ratio of 1:10 by weight.

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EXHIBIT 2

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JACOBACCI & PAKINENS SPA

NO. 8423 P. 19

Japanese Unexamined Patent Application Laid Open (Kokai) H7-102114

TRANSLATION FROM JAPANESE

(19) Japanese Patent Office (JP)

(12) Laid Open Patent Application Gazette (A)

(11) Laid Open Patent Application (Kokai) H7-102114

(43) Date Laid Open: April 18, 1995

(51) Int. Cl⁸ Recog. Code File No. FI Tech. Disp. Loc.

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LAG

C08K 3/00

C08L 3/02 LAV
29/04 LGS

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(54) Title of the Invention: Biodegradable compositions

(57) Abstract

Purpose:

To provide cellulose ester based compositions which have excellent biodegradability with which fibres, resins and films, for example, can be moulded easily.

Constitution:

A biodegradable composition which contains cellulose ester, starch and plasticizer, in which the plasticizer content in said biodegradable composition is from 30 to 70 wt% and the proportions by weight of cellulose ester and starch are from 99 : 1 to 20 : 80.

Scope of the Patent Claims**[Claim 1]**

Biodegradable composition containing cellulose ester, starch and plasticizer, characterized in that the plasticizer content in said biodegradable composition is from 30 to 70 wt% and the proportions by weight of cellulose ester and starch are from 99 : 1 to 20 : 80.

[Claim 2]

Biodegradable composition, according to Claim 1, characterized in that the biodegradable composition contains at least one type of additive selected from among the group comprising talc, calcium carbonate, magnesium carbonate, clay, silica, alumina, glass powder, kaolin, mica, wood, cellulose, poly(vinyl alcohol), chitin, chitosan, collagen, fibroin and keratin.

Detailed Explanation of the Invention**[0001]****Industrial Field of Application**

The invention concerns cellulose ester based compositions which have excellent biodegradability which can be moulded easily as fibres, resins and films, for example.

[0002]**Prior Art**

In recent years, synthetic fibres and synthetic resins and films have found increasing use in various applications in view of their cheapness and their excellent durability and mechanical strength.

[0003]

However, when these mouldings are disposed of in the natural world they remain as they are without being degraded and so cause environmental pollution, and this is becoming a serious social problem.

[0004]

In order to resolve this problem the mouldings must be capable of being biodegraded by the micro-organisms which are present in soil and water, and progress has been made with the development of compositions with which the biodegradation rate can be controlled.

[0005]

For example, synthetic resin materials in which plasticizer and polyester are mixed with cellulose ester have been disclosed in Japanese Unexamined Patent Application Laid Open H4-142344 but, as is clear from the description in lines 5 and 6 of the lower right hand column on page 4 of said specification, said synthetic resin requires a long period of time to be degraded in that "the time for a moulding of thickness 1 mm to collapse to the extent that small parts cannot be seen is from 1 to 5 years", and the biodegradability cannot be said to be adequate for preventing the occurrence of environmental pollution.

[0006]

On the other hand, biodegradable compositions where starch is compounded in ethylene - vinyl alcohol copolymers or aliphatic polyesters have been disclosed in Japanese Unexamined Patent Applications Laid Open H5-32822, H4-353537 and H4-248851.

[0007]

However, said compositions have a disadvantage in that the moulding properties and physical properties are poor because starch has poor compatibility with the ethylene - vinyl alcohol copolymers and aliphatic polyesters which are the other compounding ingredient.

[0008]

That is to say, conventionally, no resin composition which has excellent biodegradability, especially in a short period of time, and of which the moulding properties and physical properties are not adversely affected has yet been suggested.

[0009]

Problem to be Resolved by the Invention

The aim of the present invention is to resolve the problems of the prior art outlined above and to provide cellulose ester based compositions which have excellent biodegradability and with which fibres, resins and films, for example, can be formed easily.

[0010]

Means of Resolving These Problems

As a result of a thorough investigation carried out with a view to achieving the aforementioned aim of the invention, the inventors have discovered that the prescribed biodegradable compositions can be obtained with novel compositions, which are obtained using cellulose esters and a novel component known as a starch, which have excellent compatibility with cellulose esters.

[0011]

Thus there is provided by means of the present invention a biodegradable composition containing cellulose ester, starch and plasticizer which is characterized in that the plasticizer content in said biodegradable composition is from 30 to 70 wt% and the proportions by weight of cellulose ester and starch are from 99 : 1 to 20 : 80.

[0012]

The essential constitution of the invention is described in more detail below. The cellulose ester which is used in the invention is a material where some or all of the hydroxyl groups of cellulose have been substituted with ester groups, and examples include cellulose diacetate, cellulose triacetate, cellulose acetate butyrate and the like.

[0013]

These cellulose esters can be used individually, or two or more can be used conjointly.

[0014]

Furthermore, the starch which is compounded with the abovementioned cellulose esters may be a fresh starch obtained from corn, wheat, Irish potatoes, rice, tapioca, sweet

potatoes and the like, or a physically modified starch such as α -starch, an enzymatically modified starch such as dextrin or amylose, a chemically modified starch such as oxidized starch, or any other starch derivative such as an etherified starch or a crosslinked starch.

[0015]

In this invention it is important that the proportions by weight of the abovementioned cellulose ester and starch are from 99 : 1 to 20 : 80.

[0016]

In a case where the proportion by weight of cellulose ester exceeds 99 the biodegradation rate is too slow and the material is impractical, while in a case where the proportion by weight is less than 20 not only are the moulding properties affected adversely but the biodegradation rate is too high and the composition is unstable.

[0017]

The compounding proportions of the cellulose ester and starch should be set appropriately within the abovementioned range in accordance with the prescribed degree of biodegradability, and the biodegradation rate falls as the amount of cellulose ester compounded is increased.

[0018]

Furthermore, the plasticizer used in the invention should be biodegradable, and some examples are indicated below.

[0019]

(1) Organic Acid Esters

Aromatic esters such as phthalate esters, natural fats, monoglycerides, diglycerides, fatty acid esters, cyclic esters such as caprolactone for example, oligomers such as adipic acid - ethylene glycol oligomers, and the like.

(2) Sugar Esters

Sorbitol butyrate, mannitol acetate propionate and the like.

(3) Organic Acids

Saturated fatty acids such as lactic acid, citric acid, caproic acid and tartaric acid, unsaturated fatty acids such as fumaric acid and oleic acid, aromatic acids such as phthalic acid, and the like.

(4) Others

Glycols such as ethylene glycol, propylene glycol, tetramethylene glycol, triethylene glycol, polyethylene glycol and polypropylene glycol, glycerine, diglycerine, triethylenimine, triethanolamine, sulphorane, dimethoxyethyl adipate, ethyl cellosolve, methyl cellosolve, triethylene glycol acetate, caprolactam, and the like.

[0020]

Here, the amount of plasticizer used must be from 30 to 70 wt% with respect to the total weight of the composition which includes the cellulose ester, starch and plasticizer.

[0021]

The composition obtained becomes unstable if the amount of plasticizer used exceeds 70 wt%, while if the amount used is less than 30 wt% then the compatibility of the cellulose ester and the starch becomes poor and the moulding properties are adversely affected. Furthermore, the abovementioned plasticizers can be used individually, or two or more types can be used conjointly.

[0022]

In addition to the abovementioned components, at least one type of additive selected from among the group comprising talc, calcium carbonate, magnesium carbonate, clay, silica, alumina, glass powder, kaolin, mica, wood, cellulose, poly(vinyl alcohol), chitin, chitosan, collagen, fibroin and keratin may be compounded in a composition of this invention.

[0023]

Various aspects of performance, starting with mechanical properties such as strength for example, can be improved by adding of the abovementioned additives. However, if the amount of additive used is too great then the moulding properties are adversely affected and so the amount used is preferably limited to a maximum of 50 wt%.

[0024]

Stabilizers and modifying agents, for example, can also be compounded in a composition of the invention within the ranges where they do not obstruct the aim of the invention.

[0025]

The compositions of the invention obtained in this way can be moulded into any form, such as resin mouldings, films or fibres for example, since they have good moulding properties, and the mouldings obtained exhibit excellent biodegradability.

[0026]

Action

In this invention, the moulding properties of the composition are improved and excellent biodegradability is achieved by using a novel compounding composition of starch and cellulose ester.

[0027]

That is to say, the compatibility of the compounding ingredient with the compounded ingredient is improved by compounding a compounding ingredient which has a similar structure to the basic framework of the compounded ingredient and so it is possible to suppress the effect of the plasticizer to the minimum and to realize synergistically the actual characteristics of the respective structural components.

[0028]

The moulding properties with a composition of this invention are governed in the main by the cellulose ester and the biodegradability is governed in the main by the starch, and by combining the two a composition which has good moulding properties and excellent biodegradability is obtained.

[0029]

The invention is described in more practical terms below by means of illustrative examples. Moreover, the properties indicated in the illustrative examples were evaluated using the methods indicated below.

[0030]

(1) Mouldability

Pelletized compositions were spun using a melt spinning machine which had been furnished with a circular single-hole spinning die with a nozzle diameter of 0.75 mmφ with discharge at a die temperature of 220°C and monofilaments of diameter 0.4 mm were obtained, and at this time the spinability was assessed in respect of the four levels indicated below as an index of mouldability.

A: Could be spun with no difficulty at all

B: Could be spun but a slight discoloration was observed.

C: Difficult to spin with frequent thread breakage.

D: The composition degraded markedly and spinning was impossible.

[0031]

(2) Biodegradability

Monofilaments of diameter 0.4 mm, were embedded in soil collected at the Teijin K.K. Osaka Research Centre, 4-1 Mihara-3-chome, Ibaraki-shi, Osaka-fu, Japan and stored for 3 months at a temperature of 30°C with adequate humidity and the extent of degradation was then assessed on the basis of the four levels indicated below.

a: Almost completely degraded, the form of the filament being unrecognizable.

b: The form was recognizable but severe damage was apparent.

c: Part of the filament was seen to be undamaged.

d: Virtually no degradation at all was observed.

[0032]

Example 1

Cellulose diacetate flakes, Irish-potato starch and the plasticizers indicated in Table 1 were mixed in the prescribed quantities and then the mixture was milled and extruded using a screw-type extruding machine and pellets were obtained.

[0033]

The pellets so obtained were moulded in the form of monofilaments of diameter 0.4 mm using a melt spinning machine which had been furnished with a circular single hole spinning die of nozzle diameter 0.75 mmφ with discharge at a die temperature of 220°C.

[0034]

The mouldability when obtaining the monofilaments and the biodegradability of the monofilaments obtained were assessed and the results obtained are also shown in Table 1.

[0035]

Table 1

Experiment No.	1	2	3	4	5	6
Cellulose Diacetate (g)	60	40	25	15	50	50
Irish-potato Starch (g)	40	60	75	85	50	50
Plasticizer (g)						
PEG400	70	100	150	200		
Dimethyl Phthalate					25	25
Glycerine	20	20	20			25
Proportion of Plasticizer by Weight (wt%)	47	55	63	67	20	33
Milling and Extrusion Temperature (°C)	215	210	207	205	220	220
Mouldability	A	A	A	D	D	A
Biodegradability	b	a	a	-	-	b

(Experiment Numbers 4 and 5 are comparative examples)

[0036]

Example 2

The same procedure as in Example 1 was carried out except that the type of plasticizer used in Example 1 was changed in the way shown in Table 2.

[0037]

The mouldability when obtaining the monofilaments and the biodegradability of the monofilaments obtained were assessed and the results obtained are also shown in Table 2.

[0038]

Table 2

Experiment No.	7	8	9	10	11	12	13
Cellulose Diacetate (g)	50	50	50	50	50	50	60
Irish-potato Starch (g)	50	50	50	50	50	50	40
Plasticizer (g)							
Dimethyl Phthalate	25	25	25	25	25	50	
Diethyl Phthalate	25						
Diethyl Sebacate		25					70
Triacetin			25				
Sulphorane				25			
Diethyl Maleate					25		
Proportion of Plasticizer by Weight (wt%)	33	33	33	33	33	33	41
Milling and Extrusion Temperature (°C)	220	220	220	220	220	220	220
Mouldability	A	A	A	A	A	A	A

Biodegradability	c	b-c	b-c	b	b-c	c	b-c
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[0039]

Example 3

The same procedure as in Example 1 was carried out except that the additives shown in Table 3 were included in Example 1.

[0040]

The mouldability when obtaining the monofilaments and the biodegradability of the monofilaments obtained were assessed and the results obtained are also shown in Table 2.

[0041]

Table 3

Experiment No.	14	15	16	17	18	19
Cellulose Diacetate (g)	50	50	60	50	100	25
Irish-potato Starch (g)	50	50	40	50	-	75
Plasticizer (g)						
PEG400			70			200
Dimethyl Phthalate	25	25		25	40	20
Glycerine	25	25	20	25		30
Proportion of Plasticizer by Weight (wt%)	33	33	47	33	29	71
Additive (%)						
Talc	30			75		
Cellulose		30				
Water			4			
Milling and Extrusion Temperature (°C)	220	220	207	220	220	200
Mouldability	A	A	B	C	C	D
Biodegradability	a	a	b	b	d	-

(Experiment Numbers 18 and 19 are comparative examples)

[0042]

Effect of the Invention

By means of the invention there are provided cellulose ester based compositions which can be moulded easily into fibres, resins and films and which have excellent biodegradability

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EXHIBIT 3

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CALCULATION SHEET

D/E

ACCORDING TO THE PRESENT INVENTION:

P = PLASTICIZER

A = STARCH

E = CELLULOSE ESTER

$$\frac{A}{E+P} = \begin{cases} 11.6 \\ 11.8 \end{cases} \quad (3)$$

$$\frac{P}{E} = 0.1 \text{ TO } 0.4 \quad (4)$$

USING EQUATIONS (3) AND (4)
WE MAY CALCULATE THE RATIO:

$$\frac{P}{E+P+A}$$

$$\text{FROM (3): } (E+P):A = \begin{cases} 0.6:1 \\ 1.8:1 \end{cases}$$

$$\text{THEREFORE } (E+P+A):(E+P) = \begin{cases} (0.6+1):0.6 \\ (1.8+1):1.8 \end{cases}$$

$$\frac{(E+P)}{(E+P+A)} = \begin{cases} \frac{0.6}{1.6} \\ \frac{1.8}{2.8} \end{cases}$$

WHICH MAY BE WRITTEN

$$.375 \leq \frac{E+P}{E+P+A} \leq .947$$

$$\text{MULTIPLY BY } \left(\frac{P}{E+P}\right): .375 \left(\frac{P}{E+P}\right) \leq \frac{E+P}{E+P+A} \left(\frac{P}{E+P}\right) \leq .947 \left(\frac{P}{E+P}\right)$$

$$.375 \left(\frac{P}{E+P}\right) \leq \frac{1}{E+P+A} \leq .947 \left(\frac{P}{E+P}\right)$$

$$.375 \left(\frac{1}{E+P+1}\right) \leq \frac{P}{E+P+A} \leq .947 \left(\frac{1}{E+P+1}\right)$$

THEREFORE, ACCORDING TO (4)

$$\text{WHEN } \frac{P}{E} = 0.1 \Rightarrow 3.4\% \leq \frac{P}{E+P+A} \leq 8.6\%$$

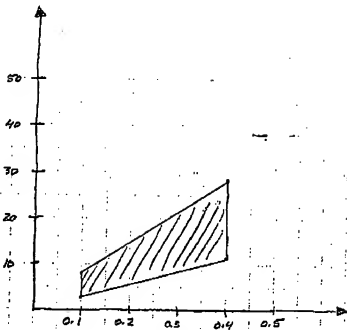
$$\text{WHEN } \frac{P}{E} = 0.4 \Rightarrow 10.7\% \leq \frac{P}{E+P+A} \leq 24\%$$

CALCULATION SHEET

②/5

IN GRAPHICAL FORM

$$\frac{P}{E+P+A} \%$$



(3)5

ACCORDING TO JP '07102114

$$\frac{P}{E+P+A} = 0.3 \text{ TO } 0.7 \quad (1)$$

$$\frac{E}{A} = \begin{matrix} 99:1 \\ 1:4 \end{matrix} \quad (2)$$

$$\text{FROM (1): } (E+P+A):P = \begin{matrix} 1:0.3 \\ 1:0.7 \end{matrix}$$

$$\text{THEREFORE } ((E+P+A)-P):P = \begin{matrix} (1-0.3):0.3 \\ (1-0.7):0.7 \end{matrix}$$

$$(E+A):P = \begin{matrix} 0.7:0.3 \\ 0.3:0.7 \end{matrix}$$

$$\frac{P}{E+A} = \begin{matrix} \frac{0.7}{0.3} = 2.33 \\ \frac{0.3}{0.7} = 0.428 \end{matrix}$$

ACCORDING TO (2):

$$\frac{E}{A} = \begin{matrix} \frac{99}{1} \\ \frac{1}{4} \end{matrix}, \quad A = \frac{E}{99}, \quad A = 4E$$

THEREFORE THE FOLLOWING FOUR EQUATIONS ARE OBTAINED:

$$\frac{P}{E + \left(\frac{E}{99}\right)} = 2.33$$

$$\frac{P}{E + (4E)} = 2.33$$

$$\frac{P}{E + \left(\frac{E}{99}\right)} = 0.428$$

$$\frac{P}{E + (4E)} = 0.428$$

(4)5

SOLVING THE EQUATIONS FOR $\left(\frac{P}{E}\right)$:

$$\frac{P}{E + \left(\frac{E}{99}\right)} = 2.33 \Rightarrow \frac{P}{E} = 2.33 \left(\frac{100}{99}\right) = 2.35 = 235\%$$

$$\frac{P}{E + \left(\frac{E}{99}\right)} = 0.428 \Rightarrow \frac{P}{E} = 0.428 \left(\frac{100}{99}\right) = 0.43 = \underline{43\%}$$

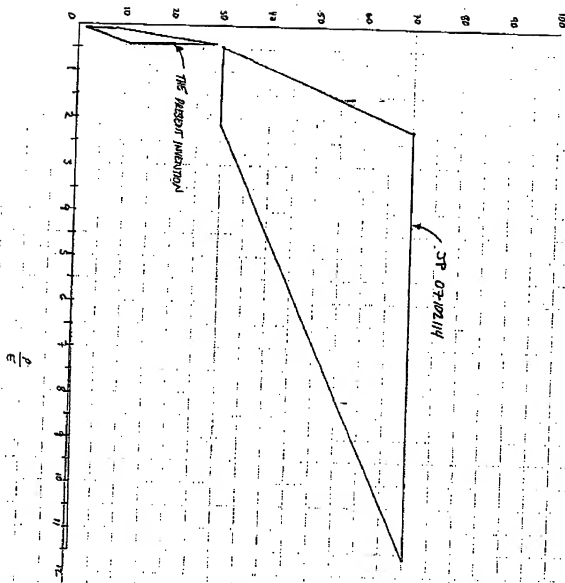
$$\frac{P}{E + (4\%)} = 2.33 \Rightarrow \frac{P}{E} = 2.33(5) = 11.65 = \underline{1165\%}$$

$$\frac{P}{E + (4\%)} = 0.428 \Rightarrow \frac{P}{E} = 0.428(5) = 2.14 = \underline{214\%}$$

$$\frac{p}{u+p+10} \%$$

⑤/5

GRAPHICAL COMPARISON OF PASSOUT INVENTION AND OF 07102119 (KOREA '19)



DEC. 9. 2002 2:31PM

NO. 8423 P. 34

EXHIBIT 4

Marked-Up Version of the Claims

--1. Biodegradable heterophase compositions comprising (1) partially or completely destructurised and/or complexed starch, (2) a polysaccharide ester, and (3) a plasticizer for the polysaccharide ester, in which the polysaccharide ester constitutes the matrix and the starch the dispersed phase,

characterized in that the compositions comprise starch and plasticized polysaccharide ester in a ratio by weight of from 1:0.6 to 1:18, the polysaccharide ester is plasticized with a plasticizer in a quantity of from 10 to 40% by weight referred to the polysaccharide ester and the starch is in the form of particles or domains of average numeral dimension [~~numeric-mean-dimension~~] lower than 1 μm for at least 80% of the particles,

the biodegradable heterophase compositions further comprising an additive which can increase and maintain a pH of 4 or more for [~~at values of 4 or more the pH of~~] a solution obtained by placing the compositions in pellet or particle form in contact with water at ambient temperature for 1 hour with the use of a pellet/particles:water ratio of 1:10 by weight.—

--2. Biodegradable compositions according to Claim 1, in which the polysaccharide ester is a cellulose ester or a starch ester.--

Inc
E1